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Asymptotic techniques based on the limit of high activation energy have been used to analyze the ignition process of a condensed fuel suddenly exposed to a hot stagnant gas. Several cases are studied differing in the location of the exothermic ignition reaction. In a first part, an analysis is presented of the gasification process when an equilibrium process or a rate process is considered to determine the gasification rate, and the use of these results to study ignition by a fast exothermic gas-phase reaction. In a second part an analysis is presented of ignition by an heterogeneous exothermic reaction including the

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) convective effects of the gasification flow. In a third part an analysis is presented of ignition by a condensed phase reaction and the derivation of a closed form expression for the ignition time. In a fourth part an analysis is presented of the heterogeneous ignition precess by convective heating described by a Newtonian law. Details of these analyses are given in previous Scientific Reports.

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GASIFICATION AND IGNITION OF A CONDENSED FUEL

BY A HOT STAGNANT GAS

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GASIFICATION AND IGNITION OF A CONDENSED FUEL BY A HOT STAGNANT GAS

M. Kindelán and A. Liñán

ABSTRACT

Asymptotic techniques based on the limit of high activation energy have been used to analyze the ignition process of a condensed fuel suddenly exposed to a hot stagnant gas. Several cases are studied differing on the location of the exothermic ignition reaction. In a first part, an analysis is presented of the gasification process when an equilibrium process or a rate process is considered to determine the gasification rate, and use these results to study ignition by a fast exothermic gas-phase reaction. In a second part an analysis is presented of ignition by an heterogeneous exothermic reaction including the convective effects of the gasification flow. In a third part an analysis is presented of ignition by a condensed phase reaction and derive a closed form expression for the ignition time. In a fourth part an analysis is presented of the heterogeneous ignition process by convective heating described by a Newtonian law.

Details of these analyses are given in previous Scientific Reports.

I. Introduction

The ignition properties of solid propellants are analyzed experimentally by exposing the propellant to a variety of heating sources, such as convective heating by hot gases, arc-image furnace heating and heating by hot stagnant gases. This last heating mechanism is established when a shock wave is reflected by a propellant placed at the end of a shock tube.

In order to relate the kinetic properties of the propellant to the results of the experimental measurements, many theoretical analyses have been developped to describe quantitatively the ignition process. See the review by Merzhanov and Averson[1].

In connection with the process of ignition by hot stagnant gases some numerical analyses have been carried out for models considering either an heterogeneous ignition reaction without gasification effects[2] or a gas phase ignition reaction including the effects of an endothermic gasification reaction, see for example Hermance et al[3].. In addition, a number of approximate analyses of the ignition processes have been carried out, aiming at simplifying the correlation of the experimental results[4-7]; these analyses do not include the convective effects due to gasification.

Under this Grant we have used asymptotic methods to generate closed form expressions relating the ignition time to the physicochemical properties of a propellant exposed to a hot oxidizing gas, with different locations of the ignition reaction, accounting for the convective effects due to gasification.

An analysis of the endothermic gasification of a condensed material by a hot gas was carried out in reference 8. The analysis can also be used to describe the ignition process when a fast exothermic reaction takes place in the gas phase immediately after gasification of the fuel.

A summary of this analysis is given in Section II.

The effects of the gasification flow in the heterogeneous ignition of a condensed fuel by a hot oxidizing gas was analyzed in reference 11. The results are summarized in Section III.

The analysis of reference 12 refers to the case when the reaction responsible for ignition takes place in the condensed phase and the solid is heated by a hot gas. A summary of the results is given in Section IV.

In all these analyses we take advantage of the fact that the activation energies of the ignition reaction is in practice large compared with the thermal energy of the solid as was done for example in references 7, 10, 13 and 14. In order to complete these asymptotic studies of ignition we carried out an analysis of heterogeneous ignition of a solid propellant subject to convective heating[15]. A summary is given in Section V.

II Endothermic Gasification

In reference 8 we develop a theoretical analysis of the gasification process of a condensed material suddenly exposed to a hot stagnant gas. Two different cases are analyzed depending on wether an equilibrium process or a rate process is considered to determine the gasification rate.

When gasification results from a rate process described by an Arrhenius law, the surface temperature, upon contact with the hot gas, rises to an intermediate "jump" value equal to the one existing in absence of gasification. Gasification however, causes the surface temperature to decrease with time, since part of the energy coming from the hot gas is absorbed by the endothermic reaction and, in addition, because the gasification flow brings cold material towards the surface and blows the hot gas away.

During an early period in which the decreasing surface regression rate is still of the order of its initial value at the jump temperature, the Arrhenius exponent can be linearized around the jump temperature and only the gradient of the non-gasification temperature profile enters the convective effects. As a result of these simplifications, the surface temperature history can be described by the solution of an integral equation involving a single parameter Δ measuring the convective effects. Closed form expressions of the temperature and concentration profiles are also given in the form of expansions for small and large values of the non-dimensional time.

For large times, when the difference between the surface temperature and the jump temperature becomes of the order of the initial temperature difference between solid and gas, the complete convective terms have to be retained and the exact form of the Arrhenius exponent has to be used. However, for those large times a quasi-similar solution exists when the distance to the surface is scaled with the square root of the nondimensional time, so that explicit expressions have been obtained for the temperature and concentration profiles.

When the gasification is assumed to obey an equilibrium Clasius-Clapeyron condition, a similarity solution exists describing the temperature and concentration profiles for all times. In this case, the surface temperature remains constant at a jump value determined by gasification effects, different, therefore, from the one existing in absence of gasification.

The analysis of the gasification process is necessary in order to study gas-phase ignition by a hot stagnant gas. In fact, the analysis presented in reference 8 may be used to describe gas-phase ignition provided that the gas-phase exothermic reaction is so fast that it occurs inmediately upon gasification. In this case, the results of the analysis may be used if the heat of gasification is replaced by the difference between the heat of gasification and the heat released by the exothermic reaction. It is found that ignition occurs only when the reaction is exothermic enough to make the convective parameter $(-\Delta)$ smaller than two.

The analyses presented in reference 8 should be extended, following the procedure of references 9 and 10 to study gas-phase ignition for reactions with large activation energies.

III Ignition by heterogeneous reaction

In reference 11 we analyze ignition of a condensed fuel by hot gases, when an exothermic heterogeneous reaction of the Arrhenius type occurs at the surface.

In the analysis we consider that the activation energy of the surface reaction is large compared with the initial thermal energy of the fuel, and we use asymptotic techniques to describe the evolution with time of the temperature and mass fraction profiles.

Previous analyses of this problem have neglected the convective effects associated with the gasification flow. In our analysis we retain these effects and show that in general they are important and, therefore, should not be neglected, without justification.

As described in Section II, the surface temperature raises, instantaneously upon contact with the hot gas, to an intermediate "jump" temperature. In absence of chemical reaction, the surface temperature remains constant and the width of the conduction layer increases with the square root of time. However, in the presence of a surface reaction there are two effects that tend to modify the temperature at the surface. First, because of the gasification flow the hot gases are blown away from the surface and cold condensed material is brought towards the surface thereby decreasing the surface temperature. Second, the energy released by the exothermic reaction has the effect of raising the surface temperature.

Two different regimes are observed depending on which of these two competitive effects dominates. If the effect of the convective flow dominates the surface temperature continuously decreases while the condensed material gasifies. The surface regression rate is maximum at the time of contact with the hot gas, and thereafter decreases with time approaching zero for long times. If the effect of heat released by the reaction dominates, the surface temperature increases with time thereby accelerating the reaction rate. This selfaccelerating behaviour causes thermal runaway of the surface temperature at a finite ignition time.

In the analysis of reference 11, the lowest order solution for the surface temperature deviation from its inert value, in an expansion in powers of the nondimensional activation energy β, is obtained by solving an integral equation. A single parameter Λ appears in this equation to measure the effects of convection. There is a critical value, $\Lambda_c=2$, that separates conditions under which ignition occurs, from conditions in which the surface temperature continuously decreases with time. The existence of these critical conditions has not been previously suggested, since this criticallity is associated with convective effects and previous analyses have neglected these effects.

For values of $\Lambda<2$, there is a thermal runaway at an ignition time $t_{ign}=t_c\ \sigma_{ign}(\Lambda)$, with the function $\sigma_{ign}(\Lambda)$ represented in figure 3 of reference 11, and with t_c given by

$$t_c = \frac{R^2 T_1^4\ (1+\Gamma)^2\ \lambda c\ \exp(2E/RT_1)}{Q^2\ Y^{2n}\ B^2\ E^2\ \rho}$$

The characteristic time is inversely proportional to the ambient oxidizer mass fraction raised to twice the reaction order of the heterogeneous reaction. The dependence on the initial gas temperature is mainly exhibited through the term $\exp(2E/RT_1)$ where T_1 is the jump temperature. For all other parameters fixed, the ignition time decreases rapidly with increasing initial gas temperature. However, when the initial gas temperature is high enough as to make the convective parameter Λ close to 2, the value σ_{ign} approaches infinity, so that the ignition time first decreases with increasing temperature and then increases to infinity.

Our analysis also shows that for large activation energy, the change in oxidizer mass fraction from its initial value is small and therefore it can be neglected when analyzing the ignition process.

Since the ignition time is strongly dependent on the jump temperature, it is important to compute correctly its value, taking into account the dependence of the thermal properties on temperature. This analysis is developped in the appendix of reference 11, and the jump temperature derived in it, is the one that should be used in all the analyses included in this Report.

IV Ignition by a condensed phase reaction

In reference 12 an analysis is presented of the ignition process of a reactive solid by conductive heating from a hot stagnant gas. To the best of our knowledge no analysis of this problem has been presented in the past. The analysis is also applicable to ignition of a reactive solid by a hot body when the thermal responsivities of both materials are of the same order.

It is found that in the limit of high activation energy of the Arrhenius exothermic reaction, the chemical heat release occurs in a thin diffusive-reactive layer near the surface. The solution for this inner layer is then matched to the solution for the outer transient-diffusive region in the solid and to that for the gas phase: the effects of the chemical reaction appears in these outer regions in the form of surface heat sources. This two-layer structure is analogous to the one found by Williams and Liñán[13].

The analysis of reference 12, for large values of the nondimensional activation energy β, leads to an integral equation to describe the evolution with time of the surface temperature deviation from its inert value. With the appropriate nondimensional variables no parameters appear in this equation so that one numerical integration is sufficient to obtain the surface temperature history and the nondimensional ignition time, $\sigma_{ign}=0.393$, when a thermal runaway is found to occur. In dimensional variables the ignition time is given in terms of the specific heat c, initial gas temperature T_{go}, jump temperature T_1, ratio of thermal responsivities Γ, pre-exponential factor A, heat released by the reaction Q and activation energy E by

$$t_{ign} = 0.393 \, \frac{c(T_{go}-T_1)\,\Gamma(1+\Gamma)}{A\,Q} \exp(E/RT_1)$$

V. Heterogeneous ignition by convective heating

Reference 15 presents an analytical description of the heterogeneous ignition of a solid under convective heating by a hot gas. The heat exchange between the gas and the solid is modelled by a Newtonian law, so that the heat flux from the gas to the solid is written as proportional to the difference in temperature of the gas and that of the surface of the solid. The exothermic heterogeneous reaction is considered to follow Arrhenius kinetics with an activation energy large compared with the thermal energy of the solid. The effects of interphase mass transfer are neglected here.

The analysis of reference 15 parallels the analysis of Niiooka and Williams [14] who studied ignition by Newtonian heating with a condensed phase reaction. In the limit of high activation energy, two ignition regimes are found. In a first regime, for large values of the frequency factor, a short reactive stage ending in thermal runaway follows a long inert stage. For small values of the frequency factor, the chemical heat release can be neglected in a first stage, but not in a much longer second stage when the surface temperature is close to the hot gas temperature; no thermal runaway or well defined ignition time exists.

Closed form expressions for the ignition time are given for both regimes.

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